

Lawrence Livermore National Laboratory

Scintillator Materials

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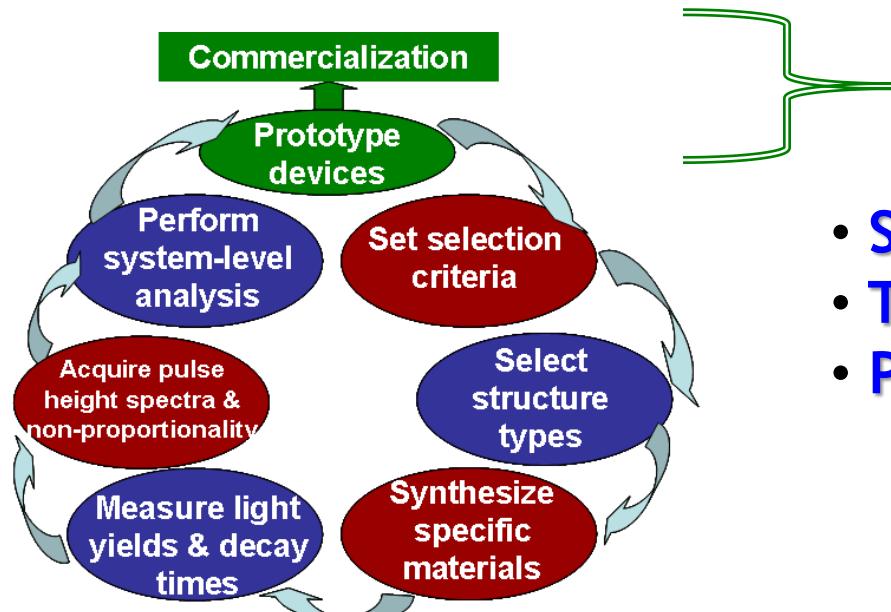
LLNL-PRES-468519

Overview

- Applications:** 1) RIBF - Accurate measurement of Doppler-shifted gammas produced in rare isotope beams during decay, fragmentation and nuclear reactions
2) CMS - Calorimetry of multi-GeV gammas

Requirements for new detector materials:

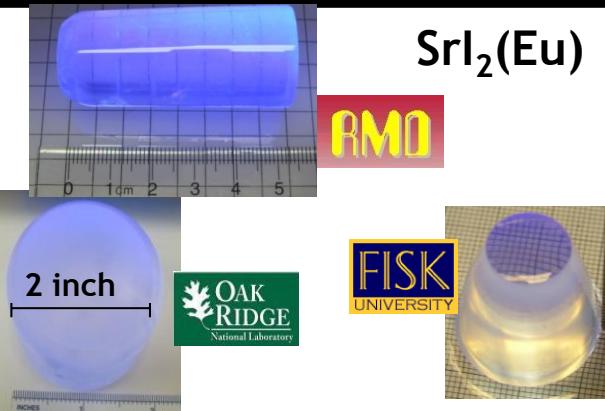
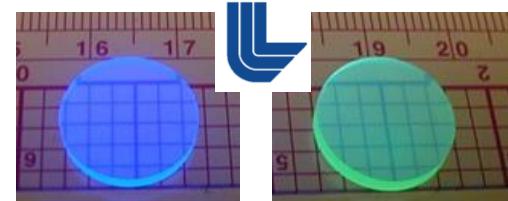
- 1) High energy resolution – to discriminate gamma spectra
- 2) High stopping – for high rate of full energy events
- 3) Fast coincidence timing w/ low dead time – observe correlated events
- 4) Radiation hardness – high rate / long duration experiments
- 5) Low cost / maintenance – starting materials, growth / ruggedness
- 6) Easy fabrication – for close-packed segmented arrays and large sizes



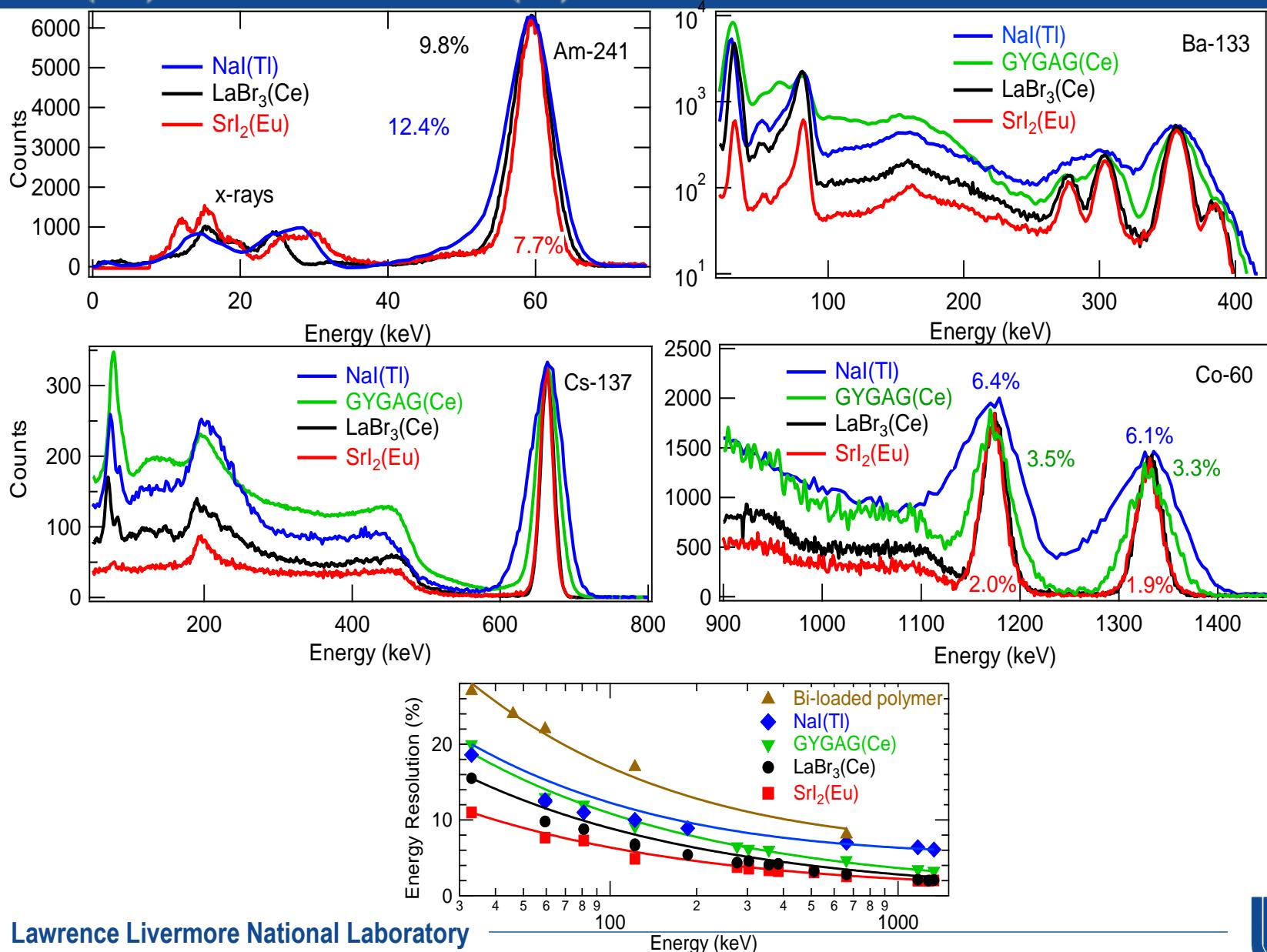
Our **Directed Search Method** has been used to discover:

- Single Crystals → $\text{SrI}_2(\text{Eu})$
- Transparent Ceramics → Garnet(Ce)
- Plastics → Bi-loaded Polymer

Inorganic single crystal and ceramic scintillators are being developed for gamma ray spectroscopy

Single crystals	Ceramics	Plastics
 <p>Srl₂(Eu) RMD FISK UNIVERSITY</p> <p>2 inch</p> 	<p>GYGAG(Ce), 1 in³</p> 	 <p>Bi-loaded polymers, 1 cm³</p>
<ul style="list-style-type: none">▪ Often hygroscopic/air-sensitive▪ Fragile/brittle▪ Complex to grow large crystals▪ Can have gradients & non-uniformity <p>▪ All crystal structures possible</p> <p>▪ Best energy resolution materials- LaBr₃(Ce), Srl₂(Eu) ~2.6% @ 662 keV</p>	<ul style="list-style-type: none">▪ Unreactive with air, water▪ Mechanically durable▪ Large sizes (100 cm³ Nd:YAG ceramics commercially available)▪ Increased activator uniformity▪ Can form high melting point oxides <p>▪ Requires cubic material</p> <p>▪ Good energy resolution- GYGAG(Ce) Gadolinium Garnet ~4.5% @ 662 keV</p>	<ul style="list-style-type: none">▪ Unreactive with air, water▪ Mechanically durable▪ Large sizes, low cost <p>▪ Non-standard polymer required</p> <p>▪ Bi-loading uniformity important</p> <p>▪ Energy resolution so far ~ 7% @ 662 keV</p>

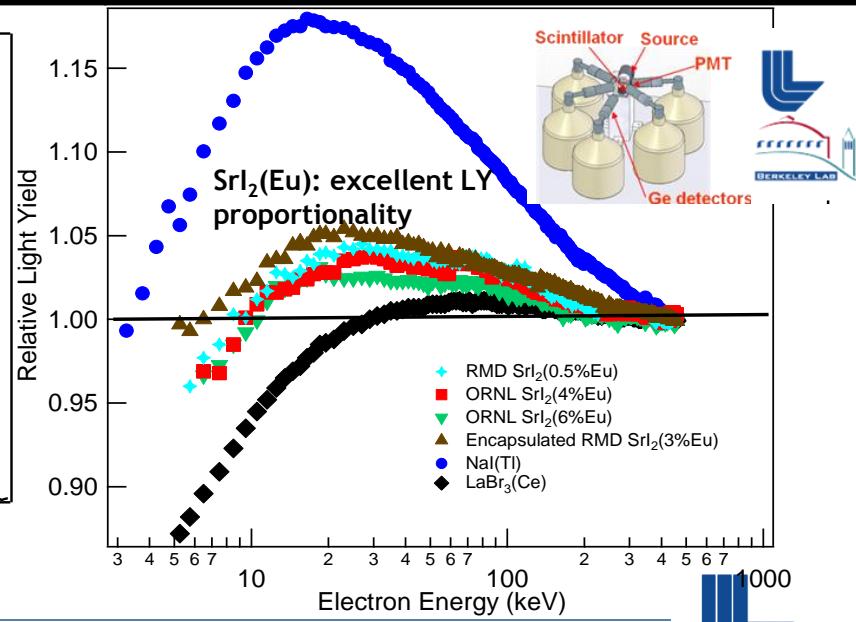
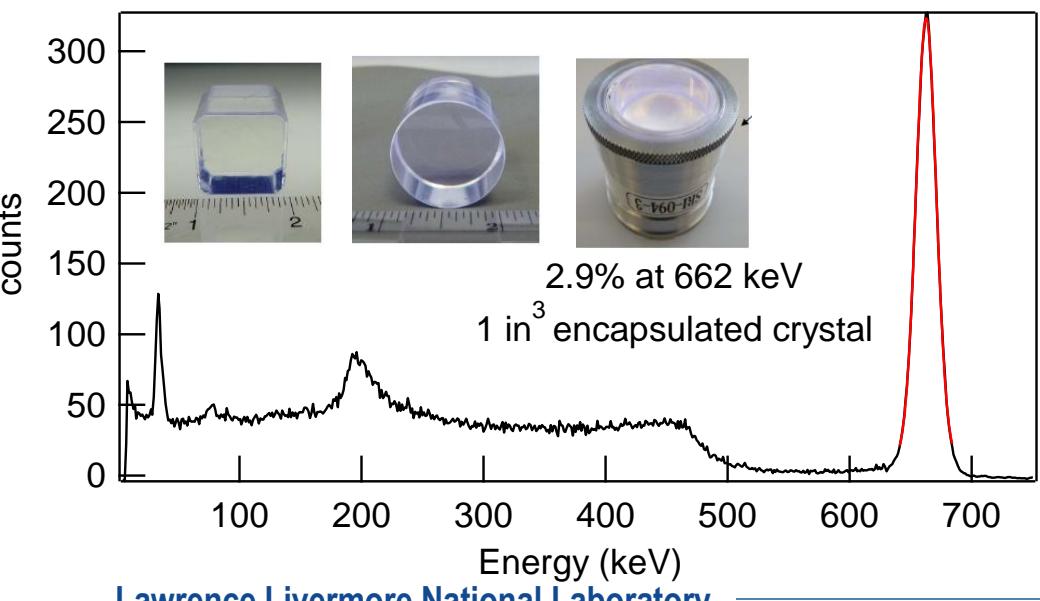
For gamma ray spectroscopy, $\text{SrI}_2(\text{Eu})$ comparable to $\text{LaBr}_3(\text{Ce})$ and GYGAG(Ce) is better than NaI(Tl)



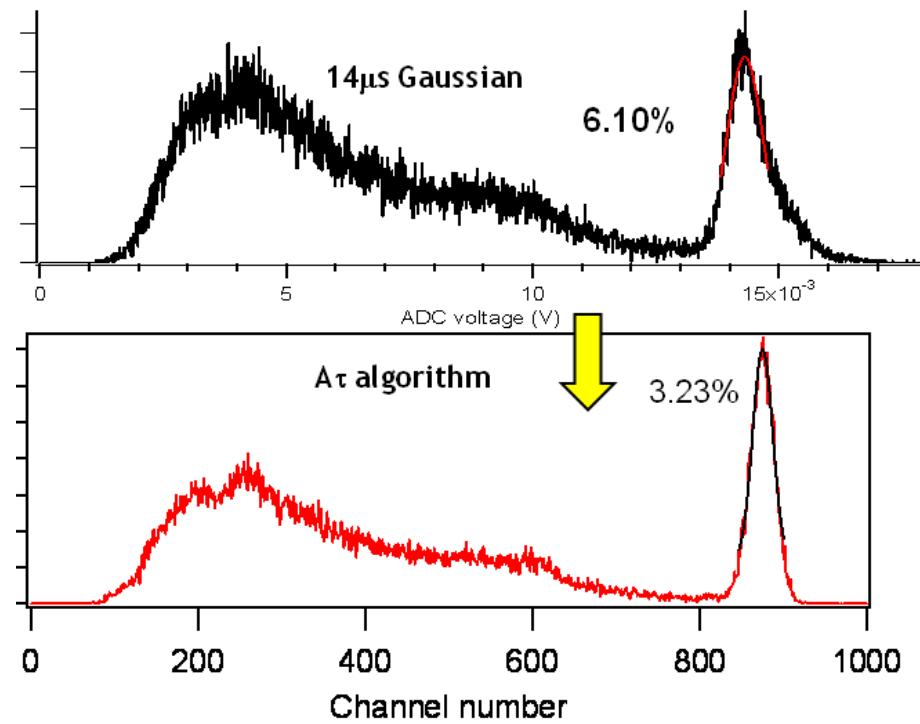
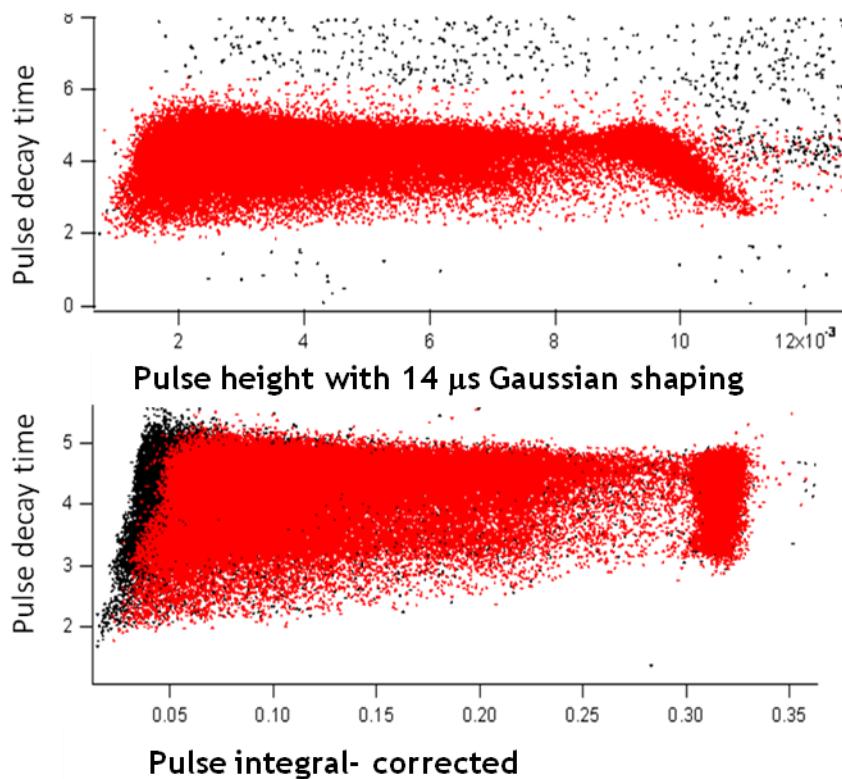
STRONTIUM IODIDE

Production of encapsulated $\text{SrI}_2(\text{Eu})$ underway

Property	$\text{LaBr}_3(\text{Ce})$	$\text{SrI}_2(\text{Eu})$	Comparison
Melting Point	783 °C	538 °C	✓ Less thermal stress
Handling	Easily cleaves	Resists cracking	✓ Better processing
Light Yield	60,000 Ph/MeV	90,000 Ph/MeV	✓ Higher
Proportionality contribution	~2.0%	~2.0%	✓ Favorable
Inhomogeneity	0%	>1% (current)	Impurities and surfaces being addressed
Decay time	30 nsec	0.5-1.5 μsec	Fast enough to avoid deleterious signal pile-up
Self-radioactivity	La ~ 3x NORM	None	✓ Less noise
Hygroscopic / air sensitive?	Very	Very	Similar
γ absorption (2x3", 662 keV)	22%	24%	Similar



Digital readout may be employed to improve energy resolution of large crystals

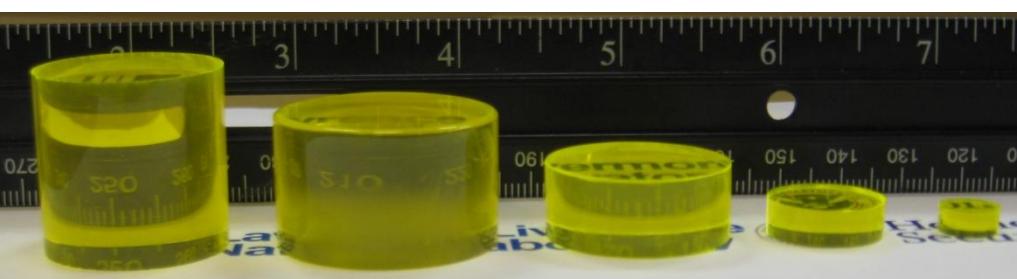


- Inverse correlation between decay time and pulse height
- Events may be corrected based on pulse shape, and energy histogram made more accurate

TRANSPARENT CERAMICS

Cubic oxides studied for transparent ceramic scintillators

Structure type	Illustrative Material	Optical properties	Activates with Ce?	Activates with Eu?
Garnet	$\text{Gd}_3\text{Sc}_2\text{Al}_3\text{O}_{12}$ - GSAG	High transparency	High LY	High LY
Perovskite	SrHfO_3 - SHO	High transparency	Modest LY	unknown
Bixbyite	Lu_2O_3	High transparency	no	High LY
Pyrochlore	$\text{La}_2\text{Hf}_2\text{O}_7$ - LHO	Moderate transparency	no	Modest LY
Eulitine	$\text{Bi}_4\text{Ge}_3\text{O}_{12}$ - BGO	Unknown	no	unknown
Defect Fluorite	Y_3TaO_7	Unknown	no	unknown
Defect Fluorite	$\text{HfO}_2\text{-Y}_2\text{O}_3$	Unknown	no	Low LY
Simple Cubic	BaO	Hygroscopic	--	--



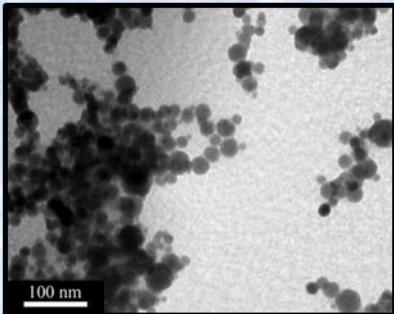
Ceramics fabrication requires multiple, optimized steps

Flame Spray Pyrolysis (Nanoparticle production)

Metal salts in organic solvent → nozzle →
Droplets ignited → Particles collected

Flame
Spray
Pyrolysis

Pratsinis, ETH

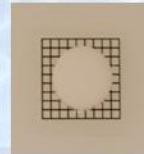


~10 nm particles

Green Body
(Powder Compact)
Slip casting
Tape casting
Pressing

Staged Heat Treatment (Calcination/Crystallization)

-Up to 1200°C in Air



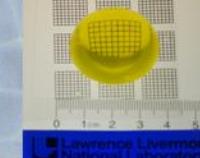
Vacuum Sintering (Densification to closed porosity)

-Up to 2000°C in 1×10^{-6} Torr Vacuum

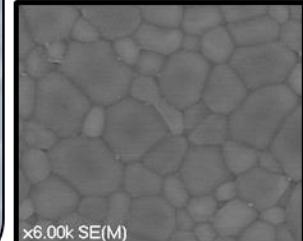


Hot Isostatic Pressing (Full Densification)

-Up to 2000°C at 29,000 psi Ar



Final grain size
~10 microns



Transparent ceramics fabrication economical for large size production of highly uniform optics

Vacuum Furnace



Hot Isostatic Press



LLNL Ceramics Facility:
Production of 10" optics

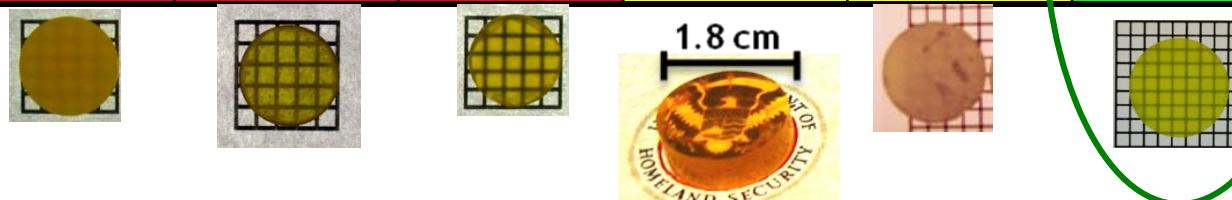
Industrial HIP, 66" diameter

Other features of ceramics:

- Cladding, layers of different materials, integral light reflectors - “as formed” in “net shape”
- Single HIP run can process many samples
- Overall, process temperatures low, rapid → completed optic in ~24 hrs
- Very high radiation hardness compared to halide single crystals
- “Unbreakable,” machinable, environmentally robust

We have been working to identify an optimal Gd-based garnet scintillator for the past five years

	2006-7	2007-8	2008-9		
Composition	GAG	GYAG	GGG	GSAG	GYSAG
Phase Stability	Poor	Moderate	Excellent	Excellent	Excellent
γ -LY(Ph/MeV)	—	40,000	—	25,000	30,000
En. Res. (662 keV)	—	11%	—	11%	10%



2010-11

Scale-up of GYGAG(Ce) and GLuGAG(Ce)

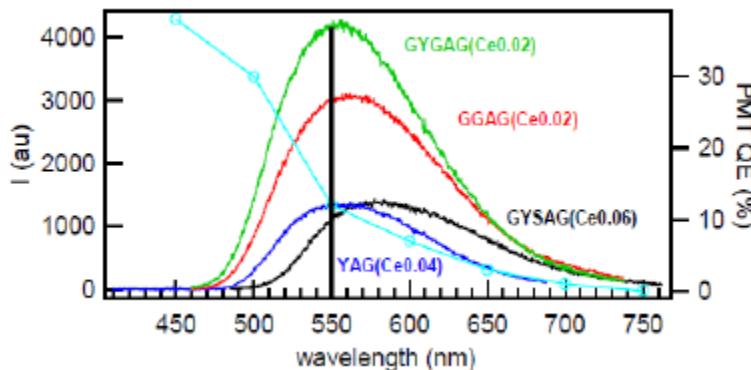


Current status:

- 1 in³ parts formed routinely

To optimize performance:

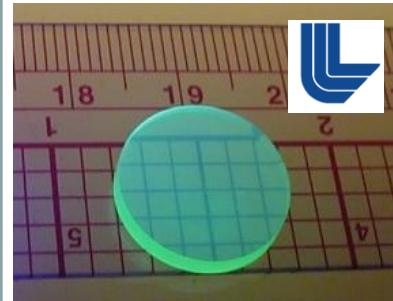
- Photodetector matched to green scintillation
- Readout for decay which includes several components



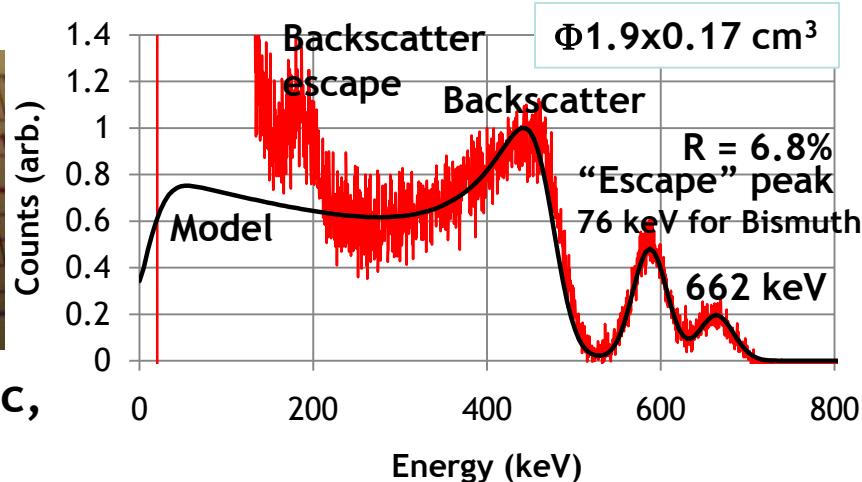
PLASTICS

Recently we have developed polymer scintillators with enhanced scintillation characteristics

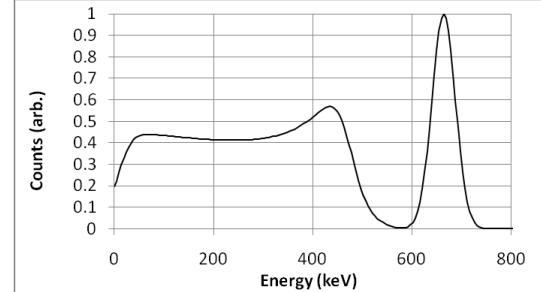
Gamma Spectroscopy Plastic: Energy resolution similar to NaI(Tl)



Bi organometallic,
40 wt%



Predicted 3" part w/ R= 8%



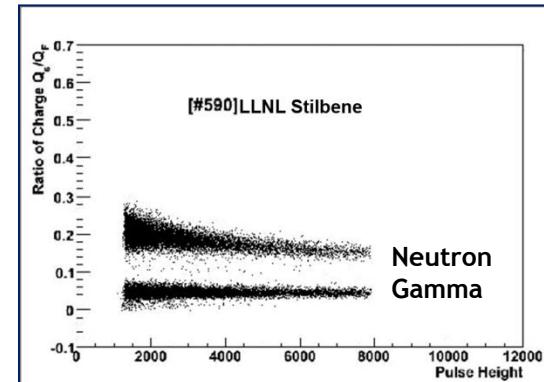
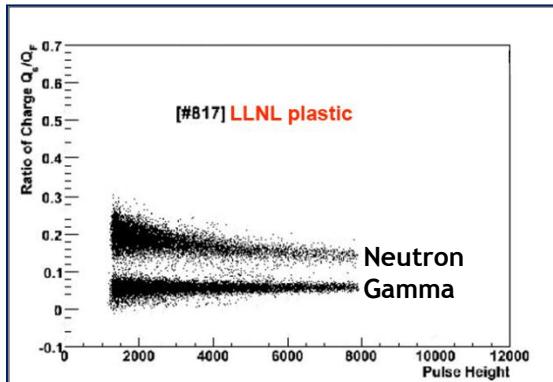
Escape peaks in small parts
eliminated when larger

Pulse Shape Discrimination “PSD” Plastic:

Neutron/gamma discrimination in new 2" PSD plastic scintillator similar to stilbene single crystal



Natalia Zaitseva and team



Materials for future gamma spectrometers?

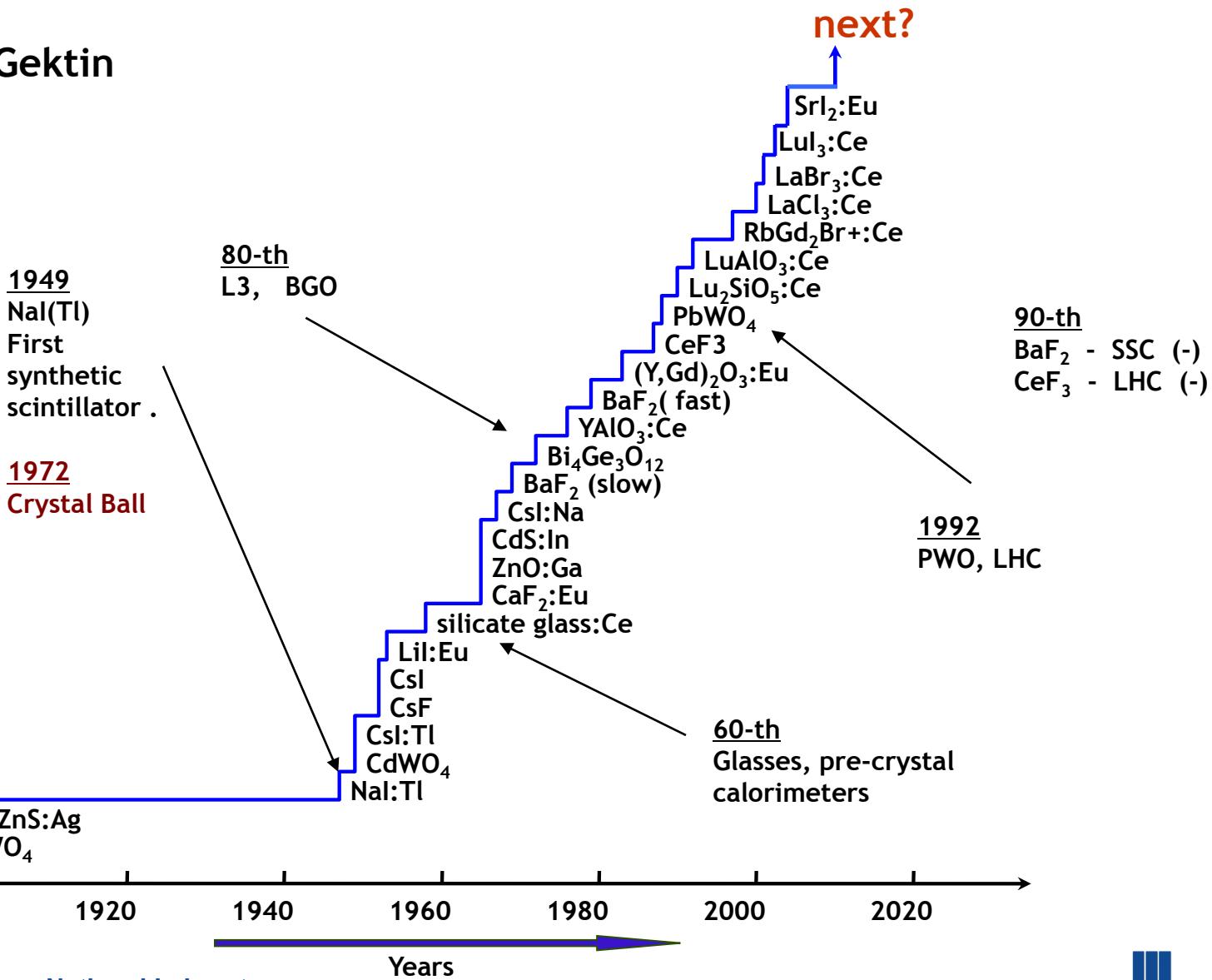
	Gamma Spectroscopy Scintillator	Z_{eff}	Light Yld (Ph/MeV)	En Res, 662 keV	Nonprop En Res, 662 keV	$S_{\text{pp}},$ 1 MeV, (15 mm x 15mm x 15 mm)
Single crystals	Nal(Tl)	50	40,000	7%	5.0%	2.2%
	LaBr ₃ (Ce)	44	63,000	3%	2.2%	2.8%
	SrI ₂ (Eu)	49	90,000	3%	2.2%	3.0%
Garnet ceramics	(Gd,Y,Lu) ₃ (Al,Ga) ₅ O ₁₂ (Ce)	47	50,000	4.5%	1.9%	3-9%
Plastics	Standard PVT	4.5	15,000	8% (Compton)	3.6%	0
	Current LLNL Bi-loaded polymers	26	10,000- 30,000	7-9%	3.5%	0.3%

Scintillator invention and use in high energy physics

Slide from A.Gektin



R. Hofstadter
1961
Nobel Prize



Crystal calorimeters

New generation of calorimeter - new scintillator development

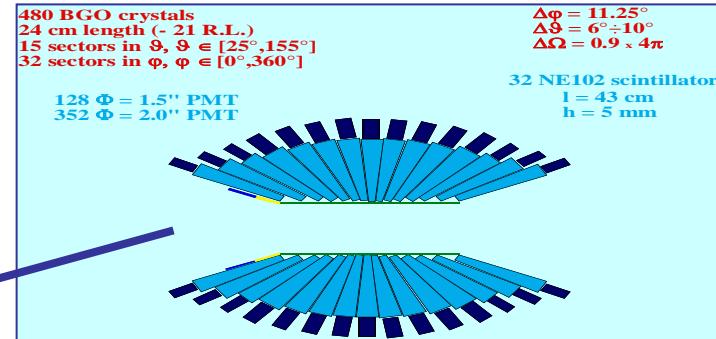
Slide from P. Lecoq

	Crystal Ball	CLEO II	L3	BaBar	Belle	L^* Lol	GEM EoI	GMS EoI	$L3P$ EoI	ALICE	CMS
Where	SPEAR	CESR	LEP	SLAC	KEK	SSC	SSC	LHC	LHC	LHC	LHC
When	1972	Late	1980's		1999						2005
Beam	$e^+ e^-$	$e^+ e^-$	$e^+ e^-$	$e^+ e^-$	$e^+ e^-$	pp	pp	pp	pp	ion ion	pp
Crystal	Nal(Tl)	CsI(Tl)	BGO	CsI(Tl)	CsI(Tl)	BaF ₂	BaF ₂	CeF ₃	CeF ₃	PbWO ₄	PbWO ₄
Number	7000	7800	11400	6800	8800	26000	15000	45000	100000	36000	82000
Length [X_0]	16	16	21.5	16	16	24.5	24.5	25	25	22	25

New!
asymmetric b-factory

Proposals

LHC



Rugby Ball

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